

# A new method of alpha ray measurement using a Quadrupole Mass Spectrometer

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## Abstract

We propose a new method of alpha( $\alpha$ )-ray measurement that detects helium atoms with a Quadrupole Mass Spectrometer(QMS). A demonstration is undertaken with a plastic-covered  $^{241}\text{Am}$   $\alpha$ -emitting source to detect  $\alpha$ -rays stopped in the capsule. We successfully detect helium atoms that diffuse out of the capsule by accumulating them for one to 20 hours in a closed chamber. The detected amount is found to be proportional to the accumulation time. Our method is applicable to probe  $\alpha$ -emitting radioactivity in bulk material.

*Key words:* Quadrupole Mass Spectrometer, Helium atoms, Counting

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## 1 Introduction

There are many ways to detect  $\alpha$  rays such as gas-flow counters and solid-state detectors[1], but all detect the corresponding ionization signal by the incident  $\alpha$  ray, instead of  $^4\text{He}$  itself. However, because  $\alpha$  particles travel only a few centimeters in the air and can be easily stopped by a piece of thin foil or paper, it is harder to detect their rays than those of beta or gamma radiation.

In this work, we suggest a new method of  $\alpha$  ray measurement that aims to detect  $^4\text{He}$  neutral atoms. If one wants to measure  $\alpha$ -emitting radioactivity in bulk material with ordinary detectors, one has to rely on  $\alpha$  rays emitted

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from the thin surface of the material because of their short range. However, many materials diffuse stopped  $\alpha$  particles out of their surface in the form of neutral helium atoms. The amount of the released helium atoms can then be measured by a Quadrupole Mass Spectrometer (QMS) in terms of the mass number  $A = 4$ . Therefore, there is the advantage of being able to measure  $\alpha$ -emitting radioactivity in bulk materials. It can be seen that a higher sensitivity for alphas can be expected.

## 2 Experimental setup and method

To examine our method of  $\alpha$ -ray measurement, we used an Amersham X.825 disc type  $^{241}\text{Am}$   $\alpha$  source (Fig. 1) and a vacuum system including a QMS. The  $^{241}\text{Am}$  source, an  $\alpha$ -emitter with a half life of  $T_{1/2} = 432.2$  y, is covered with epoxy resin 25mm in diameter and 3mm in thickness. This source emits  $3.25 \times 10^5$  alphas per second, which eventually stop in the resin and diffuse out as  $^4\text{He}$  neutral atoms. The time needed for diffusion in resin is negligible<sup>1</sup> compared to the time elapsed of about 10 years or more after the production of this  $\alpha$  source. Therefore, the helium production rate of the source is thought to be the same as that of alpha ( $3.25 \times 10^5 \text{ s}^{-1}$ ). A Quadrupole Mass Spectrometer (QMS) is often used for trace element analysis because of its high isotopic selectivity and efficiency[3]. The QMS we used is a Pfeiffer Vacuum QMS200 with a channeltron detector. The mass range is  $A = 1\text{--}100$  and the detection limit is  $1 \times 10^{-12} \text{ Pa}$  [4].

The experimental setup is shown in Figs. 2 and 3. A liquid nitrogen trap was introduced to capture unwanted out-gas especially from the resin. We controlled the valves,  $V_1$  and  $V_2$  (see Fig. 3), to measure the integrated QMS channeltron current  $Q_{\text{He}}$  of helium as follows:

- (a) Draw a vacuum to  $\sim 10^{-6}\text{Pa}$  with both valves ( $V_1, V_2$ ) open.
- (b) Keep valve  $V_1$  closed for  $T_{\text{ac}} = 1\text{h}, 4\text{h}, 10\text{h}$  and  $20\text{h}$  to accumulate helium atoms from the source. The number of helium atoms expected was  $N_{\text{He}} = 1.17 \times 10^9, 4.68 \times 10^9, 1.17 \times 10^{10}$  and  $2.34 \times 10^{10}$ , respectively.
- (c) After closing valve  $V_2$ , open valve  $V_1$  to introduce helium atoms to the QMS through an orifice for a couple of seconds.
- (d) Open valve  $V_2$  to introduce most of the remaining atoms at one time for helium detection.

In our QMS system, the bypass valve  $V_2$  and the orifice( $\phi 0.3\text{mm}$ ) are placed for a high pressure sample gas. Here, we utilized them in step (c) to avoid a

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<sup>1</sup> Diffusion time of helium through various samples of epoxy resin adhesives is found in ref [2].

drastic change in the channeltron current measured by the QMS, which was observed with just  $V_1$  open after step (b). Considering the conductance of the orifice, the amount of helium atoms lost in step (c) is estimated to be less than 2%, so we measured the integration of the channeltron currents for 5 seconds after opening valve  $V_2$  in step (d). The integration time of 5s was determined after considering the time needed for opening valve  $V_2$  by hand ( $\sim 1$ s) and the evacuation time  $T_0 \sim 0.3$ s ( $\ll 5$ s) expected by the pumping speed of our vacuum system.

To estimate the amount of spurious signal caused by the out-gas from the resin, we also measured channeltron currents against a  $^{57}\text{Co}$  source as a control covered with the same epoxy capsule as the  $^{241}\text{Am}$  source. Assuming that both sources are emitting out-gas of similar composition and amount, the net integrated channeltron current  $Q_{\text{He}}$  can be defined as the difference between the integrated current  $Q_{\text{Am}}$  and  $Q_{\text{Co}}$  with the  $^{241}\text{Am}$  and the  $^{57}\text{Co}$  source, respectively, under the same accumulation time  $T_{\text{ac}}$ . In terms of  $N_{\text{He}}$  described above,  $Q_{\text{He}}$  can be written as

$$Q_{\text{He}} = Q_{\text{Am}} - Q_{\text{Co}} = eGR_iN_{\text{He}}, \quad (1)$$

where  $e$  is the elementary charge,  $G \sim 7 \times 10^3$  is the amplification factor of the channeltron, and  $R_i$  is the detection efficiency of the QMS, i.e., the ratio of the number of helium atoms detected by the QMS compared to the initial quantity  $N_{\text{He}}$ . Note that  $N_{\text{He}}$  and  $Q_{\text{He}}$  are proportional to the accumulation time  $T_{\text{ac}}$ .

### 3 Result and analysis

We measured a set of  $Q_{\text{Am}}$  and  $Q_{\text{Co}}$  twice for each  $T_{\text{ac}} = 1\text{h}, 4\text{h}, 10\text{h}$  and  $20\text{h}$ . Fig. 4 shows a few examples of the experimental result for  $T_{\text{ac}} = 20\text{h}, 10\text{h}$  and  $4\text{h}$ . A clear difference between two sources can be seen for the data of  $T_{\text{ac}} = 20\text{h}$  and  $10\text{h}$ . The difference can also be seen in Fig. 4-(C) ( $T_{\text{ac}} = 4\text{h}$ ), though a drastic change in the channeltron current makes it unclear.

Fig. 5 shows the relationship between the accumulation time  $T_{\text{ac}}$  and the net amount of helium  $Q_{\text{He}} = Q_{\text{Am}} - Q_{\text{Co}}$  with the best fit under the assumption of linearity. Here, because of the difficulty in evaluating the exact error of  $Q_{\text{He}}$ , we estimated it at  $Q_{\text{Co}}$  for each data point, which may be a conservative overestimation. The unclarity for the data of  $T_{\text{ac}} = 4\text{h}$  described above is reflected in the error bars in this figure.

The best-fit proportionality coefficient  $(1.9 \pm 0.2) \times 10^{-12} [\text{C/h}]$  corresponds to  $R_i \sim 2 \times 10^{-6}$  according to Eq.(1). We can see certain degree of linearity

between  $T_{ac}$  and  $Q_{He}$ , but the obtained detection efficiency  $R_i$  was rather small, probably because most of the helium atoms were evacuated by the vacuum pump before they got ionized.

To estimate the ultimate detection limit under this system apart from the influence of the out-gas coming from the epoxy capsule, we measured the distribution of  $Q_{BG}$ , the background channeltron current without any sources integrated for 5 seconds, as shown in Fig. 6. The best fit to the data by the normal distribution is also shown in the figure. In this way, the standard deviation  $\sigma = 9.3 \times 10^{-14}$  [C] was obtained. We defined the detection limit  $Q_{limit}$  as

$$Q_{limit} = (1.645\sigma) \times \sqrt{2} = 2.2 \times 10^{-13} \text{ [C]} \quad (2)$$

at 95% confidence level. Here, the factor  $\sqrt{2}$  is introduced to take into account the two sets of the data, the  $^{241}\text{Am}$  and the  $^{57}\text{Co}$  source, that were used for detecting the helium signal. The obtained  $Q_{limit}$  corresponds to  $\sim 10^8$  helium atoms under our system.

Although we did not try to obtain higher detection efficiency  $R_i$  with this measurement, since the valves were controlled manually, there are some possible solutions to improve  $R_i$ :

- Atom buncher [5]

An atom buncher is a cold trap to capture target gas atoms. It consists of a metal surface that often is cooled by liquid helium. Some kinds of laser can be used to temporarily heat a spot on the surface to evaporate gas atoms. The detection efficiency  $R_i$  would improve if the heated area is placed close to the ionization chamber of the QMS. This device can be applied to the detection of helium by cooling the surface to a temperature low enough to trap the helium atoms.

- Pulsed Supersonic Valve [6]

The Pulsed Supersonic Valve(PSV) is an electromagnetic device to generate supersonic free gas jet. This valve consists of two parallel metallic plates as a gate for the sample gas. The gas is only allowed to be temporarily introduced when the gate is opened by electromagnetic repulsion between these two plates. The jet of sample gas is then injected into the ionization chamber. Similar to the atom buncher described above, higher  $R_i$  can be achieved by locating the PSV near the ionization region of the QMS.

Attaching devices such as an atom buncher and a PSV to improve detection efficiency and so obtain a higher  $R_i$ , much smaller quantities of helium atoms could be detected than under our present system. Also, some QMS are said to be able to count target atoms one by one[4], so our system could theoretically

be improved to detect a single helium atom; that is, each  $\alpha$  ray regardless of its energy if the detection efficiency  $R_i$  gets close to one.

Our method can be applied to the accurate estimations of  $\alpha$ -induced soft errors in very-large-scale integrated circuit (VLSI). Soft errors are caused by  $\alpha$  rays from a minute amount of radioactive substance in LSI packages, and have become a serious problem in VLSI circuits[7]. Using mass analysis in vacuum may enable us to reduce the detection limit of  $\alpha$ -emitting radioactivity in the packages compared to the conventional method with a gas flow proportional counter.

## 4 Conclusion

We proposed a new method for alpha( $\alpha$ ) ray measurement by detecting helium atoms with a QMS. An  $^{241}\text{Am}$   $\alpha$  source and a  $^{57}\text{Co}$  source as a control were used to examine our method. The result showed that we could successfully detect helium atoms, but the detection efficiency was only  $2 \times 10^{-6}$  under our system. Additional devices such as an atom buncher and a PSV to improve detection efficiency may allow us to reduce the detection limit. This detector cannot measure the energy of the  $\alpha$  particles, but that feature is reasonably common to many other conventional  $\alpha$  detectors and is not a shortcoming in most applications. Our method may become practicable to choose low  $\alpha$ -active material for LSI packages, which is essential to reduce  $\alpha$ -induced soft error rates in VLSI circuits.

## References

- [1] Kai Siegbahn, Alpha-, beta- and gamma-ray spectroscopy, Amsterdam, 1965.
- [2] A. Gerlach, W. Keller, J. Schulz and K. Schumacher, Microsystem Technologies **7** (2001) 17.
- [3] N. Trautmann, G. Passler and K. Wendt, Anal. Bioanal. Chem. **378** (2004) 348.
- [4] Pfeiffer Vacuum catalog: Mass spectrometer, 2005.
- [5] G. S. Hurst, et al., J. Appl. Phys. **55** (1984) 1278.
- [6] H. Pauly, Atom, Molecule, and Cluster Beams I, Springer, 2000.
- [7] Y. Tosaka, et al., Jpn. J. Appl. Phys. **45(4B)** (2006) 3185.



Fig. 1. Plastic-covered  $^{241}\text{Am}$   $\alpha$  source. The central “point” is  $^{241}\text{Am}$ .

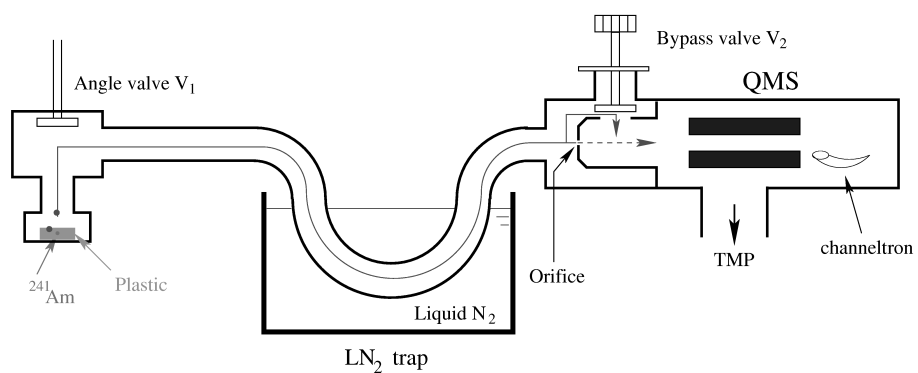


Fig. 2. Schematic view of our detection method.

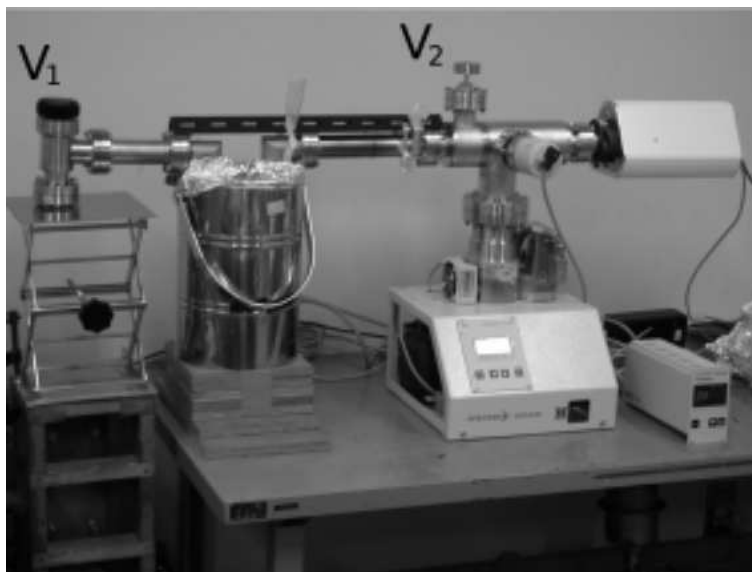
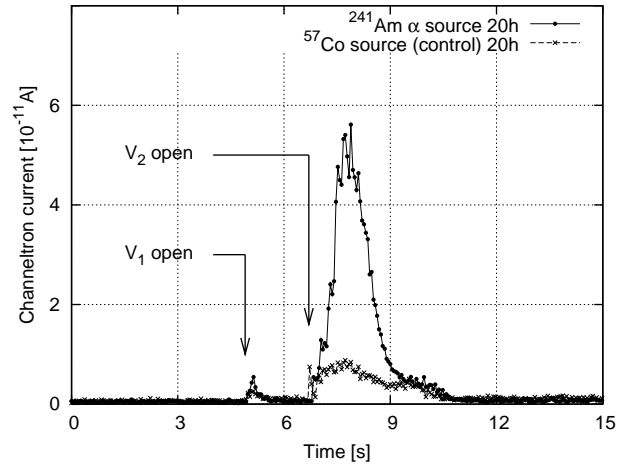
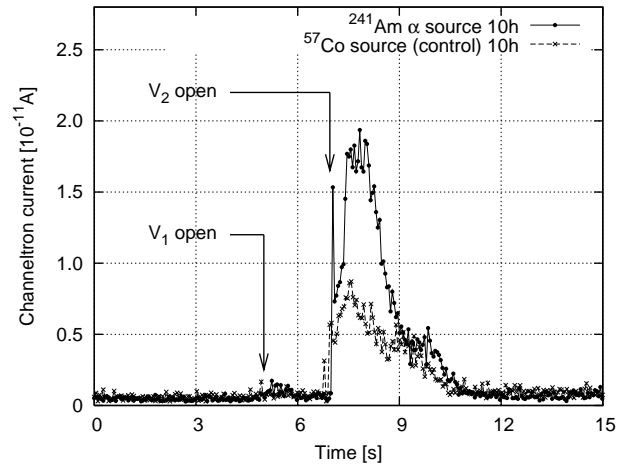


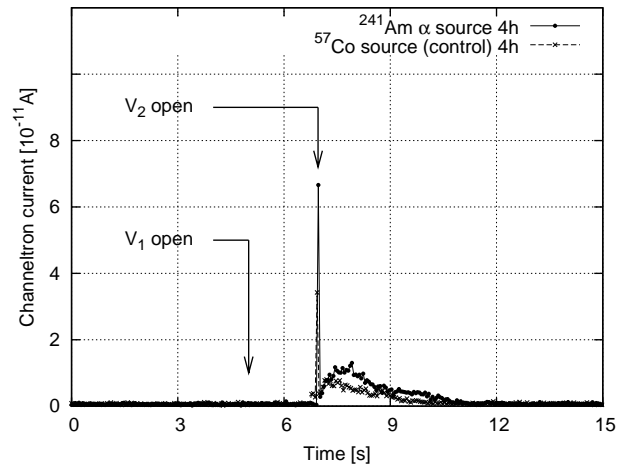
Fig. 3. Photograph of the experimental setup.



(A)  $T_{ac} = 20h$ .



(B)  $T_{ac} = 10h$ .



(C)  $T_{ac} = 4h$ .

Fig. 4. A few examples of the experimental result.

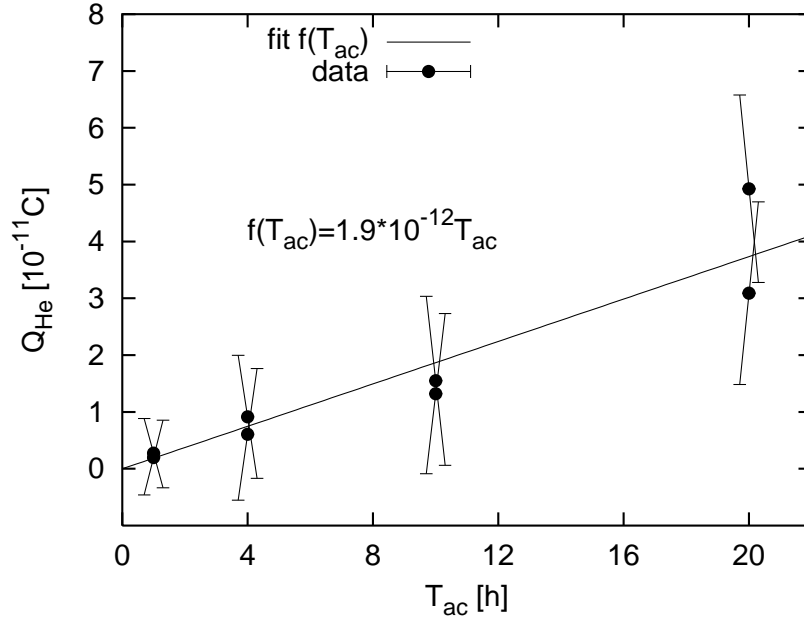


Fig. 5. Linearity between  $T_{ac}$  and  $Q_{He}$  ( $T_{ac} = 1h, 4h, 10h, 20h$ ).

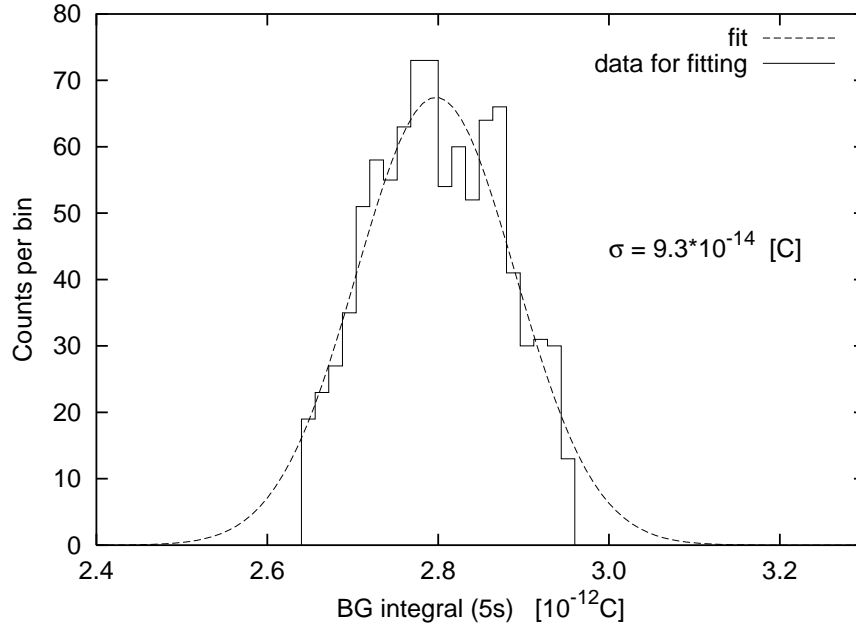


Fig. 6. Distribution of  $Q_{BG}$ .